Motion of a sphere through an aging system

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Abstract

We have investigated the drag on a sphere falling through a clay suspension that has a yield stress and exhibits rheological aging. The drag force increases with both speed and the rest time between preparation of the system and the start of the experiment, but there exists a nonzero minimum speed below which steady motion is not possible. We find that only a very thin layer of material around the sphere is fluidized when it moves, while the rest of suspension is deformed elastically. This is in marked contrast to what is found for yield-stress fluids that do not age.

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Pasty materials and concentrated suspensions have microscopic internal structure which gives them the ability to resist shear. Consequently, they can behave as soft solids or shear-thinning fluids, depending on the stress applied [1]. When the force F acting on an object within the material is less than a critical value F_c, the material responds as a solid and the object will not move. When $F > F_c$, the material around the object becomes fluidized and the object moves [2, 3]. Pasty materials typically exhibit rheological aging [4], that is, their rheological properties change with time due to slow evolution of the microstructure. The dynamics of this evolution is similar to that of glasses [4, 5], and in some cases these materials can undergo a fluid-solid transition as they age. They can also easily develop strain heterogeneities such as shear banding [6].

The motion of particles through pasty materials is important in many applications [1], and drag has been studied to some extent in yield-stress systems that do not age [2, 3, 7, 8]. The drag force on a sphere of radius R moving through a yield-stress fluid is given by

$$F = -\int_{S_c} (\mathbf{T} \cdot \hat{e}) \cdot \hat{x} \, ds. \tag{1}$$

$\mathbf{T}$ is the stress tensor and S_c the material surface on which the yielding criterion is met [1]; S_c separates the solid and liquid regions in the material surrounding the sphere. $\hat{e}$ is the outward-pointing unit vector at S_c and $\hat{x}$ a unit vector in the direction of the motion. In simple shear, the stress τ and shear rate $\dot{\gamma}$ *without* aging can be related by the Herschel-Bulkley model, $\tau/\tau_c = 1 + (K/\tau_c)\,\dot{\gamma}^n$, where τ_c is the yield stress and K and n are material parameters. The drag force can be written similarly [8]:

$$F/F_c = 1 + (K/\tau_c)\,\dot{\gamma}_{app}^n \tag{2}$$

for $F > F_c$. F_c, the minimum force required for steady motion in the liquid regime, has been calculated [2] to be

$$F_c = 4\pi R^2 \tau_c k_c \tag{3}$$

with $k_c = 3.5$. The apparent shear rate $\dot{\gamma}_{app} = v/\ell$, where v is the sphere's speed and $\ell = 1.35R$ for $n = 0.5$ [8]. With these values for k_c and ℓ, Eq. (1) indicates that, in the absence of aging, S_c is approximately R from the sphere's surface. Experiments with a non-aging yield-stress fluid agreed well with these theoretical results [3]. The effect of aging on drag has received little attention, however [9], and remains poorly understood from both the macroscopic and microscopic points of view.

Here we study the drag on a weighted ping-pong ball of diameter $2R = 3.96$ cm falling through a clay suspension that exhibits rheological aging. Since the sphere falls through previously undisturbed material, its motion at a time t probes the properties of the suspension at that time and provides a direct measure of the changes in properties as the suspension ages. We find that the drag force increases with both v and the waiting time t_w between preparation of the sample and the start of the experiment, and that v approaches a non-zero value as F approaches the critical force F_c. We show that the sphere moves through the material by fluidizing a very thin layer of the suspension — much smaller than R — immediately surrounding it, while the rest of suspension is deformed elastically. This is very different from the situation in non-aging materials [3].

We worked with Laponite RD [10], a synthetic clay known to exhibit aging [11]. Laponite consists of disk-shaped particles 30 nm in diameter and 1 nm thick. We mixed the clay 3% by weight with deionized water, adding NaOH to raise the pH to 10. The resulting suspension, with density $\rho_l = 1012$ kg/m^3, was then stored in a sealed container for four weeks to ensure complete hydration of the clay particles. Thorough remixing before each experiment was critical for obtaining reproducible results. A combination of local and large-scale mixing effectively destroyed the suspension's microstructure and put it in a reproducible initial state. After remixing the material was left to age for a waiting time t_w, during which its microscopic structure partially reformed, then the experiment was started.

The ping-pong ball had a small hole cut into it, and its density ρ_s was changed by gluing small steel beads inside it. Apart from a small piece of tape at the top to cover the hole, its surface roughness was on the order of 10 μm. This is much larger than the particle size, so slip at the sphere's surface is not expected to be important. It was dropped into the suspension from 1 cm above the free surface and its position recorded at up to 250 frames per second. The experimental container was square in cross-section, with width $L = 20$ cm and depth 45 cm. For our value of $2R/L = 0.2$, wall effects are small [7].

After an early-time regime dominated by inertia, the sphere either slows to a steady terminal velocity or stops, as shown in the inset to Fig. 1. We focus on later times, when inertial effects can be neglected. Figure 1 shows the sphere's penetration depth D (scaled by R) as a function of time t for different values of ρ_s and a fixed t_w [12]. We can distinguish three regimes [9], depending on the net gravitational force $F \propto \Delta\rho = \rho_s - \rho_l$ on the falling sphere. For sufficiently large $\Delta\rho$ (Regime 1), the sphere reaches a steady-state

speed as gravity becomes balanced by drag. For small $\Delta\rho$ (Regime 3), the speed of the sphere decreases to zero while the sphere moves a distance of order R. In the intermediate regime (Regime 2), the initial velocity v_i persists up to a displacement much larger than R, after which the sphere again slows and stops. The same three regimes are observed as t_w is increased for fixed $\Delta\rho$, as shown in Fig. 2.

Fig. 2 shows that v_i decreases with increasing t_w for a fixed $\Delta\rho$. The instantaneous speed v also decreases with t in Regimes 2 and 3, as the material continues to age. This tempts one to describe the material as having an apparent viscosity which increases with time as structure in the suspension redevelops. This naive model is inconsistent with the data, however: For both of the two largest t_w values plotted in Fig. 2, the sphere starts to slow significantly after about 100 s, much shorter than the 5 min difference in t_w between these two trials. If the stoppage were due to an increase in viscosity, this viscosity would have to increase much more over the 100 s of motion than over the additional 5 min of aging. This contradiction shows that a closer look is required.

Our results in fact indicate that the state of the material and the flow around the moving sphere are very different in the three regimes. The overall deformation γ of the suspension in Regime 3 is of order $2R/L \approx 0.2$, typical of the critical deformation γ_c at which pasty materials undergo the transition from solid to liquid [13]. This suggests that our suspension remains a solid in Regime 3. In Regime 1, in contrast, $\gamma \gg \gamma_c$ and the material around the sphere is quickly fluidized. It is also initially fluidized in Regime 2, but here the material structure redevelops on a time scale similar to that of the flow, leading to a transition from liquid to solid over the course of the run. This description is consistent with, for example, the data for $t_w = 30$ (in Regime 2) and 40 min (in Regime 3). In the former case, the sphere stops after about 600 s because the material has changed from liquid to solid, while in the latter case the material is solid at the start of the run. The material becomes solid at about the same age — approximately 40 min — in both cases. In Regimes 1 and 2, v_i depends on the state of development of the microstructure, and so on t_w. v_i is not zero in Regime 3, however, as seen in Figs. 1 and 2, so there is a minimum speed $v_c \sim 1$ mm/s that can be reached in the liquid regime. If $v_i < v_c$, the material is in its solid regime, steady motion of the sphere is not possible, and it comes quickly to a stop [14].

The rheological properties of our suspension were measured with an ARES RHS controlled-strain rheometer, using a 50 mm parallel-plate tool (plate separation 1.5 mm)

covered with sandpaper to prevent slip. Following a preshear to rejuvenate the sample, we measured the stress τ as a function of $\dot{\gamma}$. We started from high $\dot{\gamma}$ and worked downwards to limit the effects of aging on the measurements, and waited 15 s at each value of $\dot{\gamma}$. The resulting flow curve, shown by the crosses in Fig. 3, has a minimum at $\dot{\gamma}_c \approx 20$ s^{-1}, possibly indicating the presence of shear-banding at lower $\dot{\gamma}$ [15]. We take only the data for $\dot{\gamma} > \dot{\gamma}_c$ as reflecting the behavior of the homogeneous material. The instability of uniform shear flow below γ_c may be analogous to our observation that steady motion of the falling sphere is not possible for $v < v_c$, suggesting that flow instabilities analogous to shear-banding are not restricted to simple viscometric flows.

The elastic modulus $G'(t_w)$ was measured by applying an oscillatory shear with angular frequency $\omega = 1$ rad/s and deformation amplitude 5%. G' was independent of ω for $0.1 < \omega < 100$ rad/s. We found G' to increase significantly with t_w. Taking the material to be viscoelastic in the solid regime, the yield stress is given by $\tau_c = G'\gamma_c$. Using the $t_w = 0$ limit of G' and $\gamma_c = 0.2$, we find $\tau_{c,0} = 34$ Pa, consistent with the value estimated from the minimum in the flow curve. The stress data in Fig. 3 have been normalized by this value. $\tau_c(t_w)$ was estimated from $G'(t_w)$ in the same way and is plotted in the inset to Fig. 3. The yield stress increases smoothly with t_w with a logarithmic slope that increases as the material ages.

We can describe the net force F on our sphere by

$$F/F_c(t_w) = a + b\dot{\gamma}_{app}^n \tag{4}$$

for $F > F_c$. This is similar to Eq. (2) [7], but here F_c depends on t_w and a and b are treated as parameters. A fit to our falling-sphere data gives $a = 0.93$, less than the value of 1 expected for non-aging yield-stress fluids. Thus the shear rate, and so the speed v of the sphere, is greater than zero when $F = F_c$, as discussed above. F/F_c is plotted as a function of $\dot{\gamma}_{app}$ for two values of t_w in Fig. 3. To make these data consistent with the rheometric flow curve, we had to take $\ell = 0.009R$ in the calculation of $\dot{\gamma}_{app}$, a factor of 100 smaller than for non-aging yield-stress fluids. This suggests that the sheared region around the moving sphere is very thin for our aging material.

Using Eq. (3) and the data in Fig. 2, we can determine $F_c(t_w)$ for a given $\Delta\rho$. We then calculate the corresponding yield stress using Eq. (3). We again find that the yield stress increases with t_w, but to get agreement between the values of τ_c determined from

the falling-sphere data and those obtained from the measurements of G', we are forced to take $k_c = 1.085$ in Eq. (3). This is much less than the value for non-aging yield-stress fluids, and close to what is calculated from Eq. (1) if S_c coincides with the surface of the sphere [16]. This is further evidence that the moving sphere fluidizes only a thin layer of the suspension, with the lowness of k_c resulting from the fact that less force is required to disrupt the structure of a smaller volume of material. We emphasize that the low values of k_c and ℓ do not depend on any choice of constitutive relation, but are required to make the falling-sphere data consistent with the rheometric measurements.

The thinness of the fluidized layer for an aging suspension contrasts markedly with the situation in non-aging yield-stress fluids, for which the fluidized region extends approximately R from the surface of the sphere [2, 8]. This is expected to result in significant irreversible flow in the material ahead of the sphere in the latter case, but not in the former. We confirmed this by tracking the motion of 360 μm glass beads suspended in the material and illuminated by a laser sheet. Figure 4 shows the displacement of an initially horizontal line of tracer particles due to the slow passage of the sphere in a Laponite suspension and in a non-aging Carbopol gel [3] (here $F/F_c = 1.5$). The displacement calculated for a Newtonian fluid [17] is also shown. The amplitude of the deformation is much smaller in the Laponite suspension, and does not change significantly as the nominal shear rate v/R is varied from 1 s^{-1} to 30 s^{-1}. The inset to Fig. 4 shows the trajectory of a particle initially located about $R/3$ from a vertical line through the center of the sphere in the Laponite suspension. This particle moves horizontally to let the sphere pass by then nearly returns to its initial position, but is not displaced significantly in the vertical direction.

We have shown that the fluidized region around a moving sphere is approximately two orders of magnitude thinner in a suspension which exhibits rheological aging than in a non-aging material. This behavior appears to be related to the shear-banding instability observed in such materials in simple shear, but the flow in the present case is more complex. Our results are relevant to understanding the motion of objects in yield-stress materials. While we studied the motion of a macroscopic sphere, similar behavior may occur on much smaller scales around microscopic particles diffusing within the suspension [18], or even around the colloidal particles that make up the suspension itself, since the stresses involved are of the same order of magnitude.

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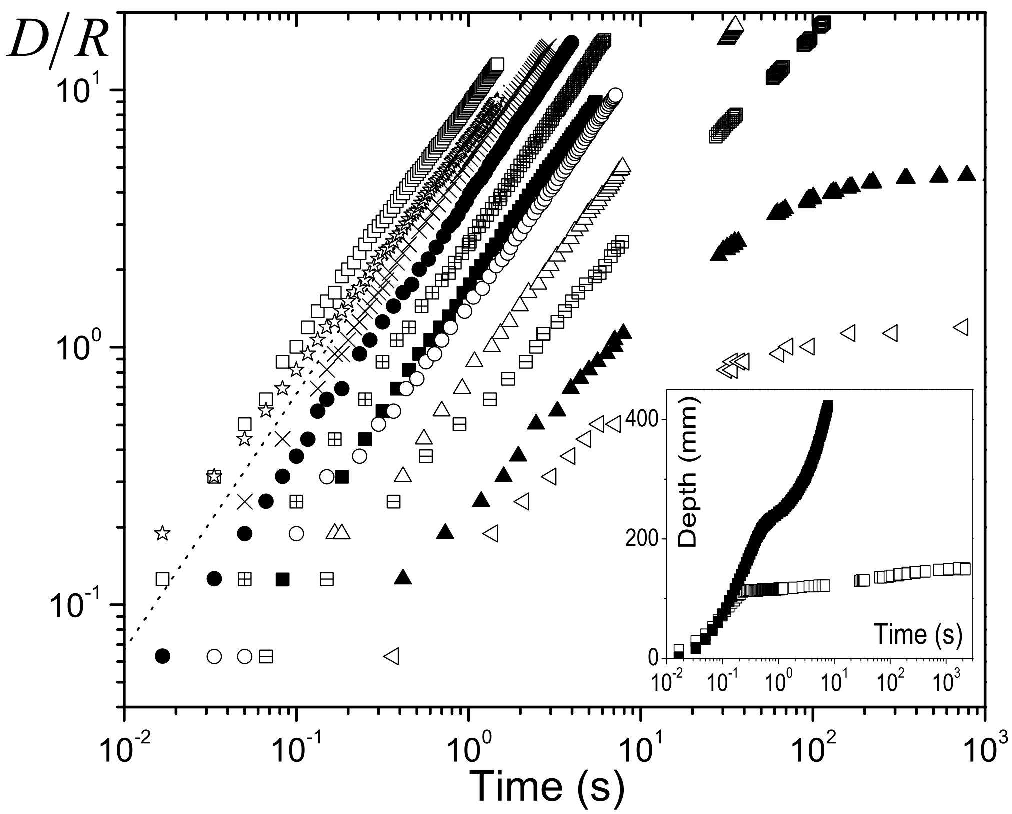

FIG. 1: Scaled penetration depth D/R vs. time t for the sphere falling through the Laponite suspension for, from left to right, $\Delta\rho = 1589$, 1524, 1460, 1389, 1319, 1251, 1153, 1057, 956, and 890 kg/m^3. Here $t_w = 4$ min. The dotted line has a logarithmic slope of 1, corresponding to a constant speed. The inset shows D as a function of t for $t_w = 20$ min and $\Delta\rho = 1806$ kg/m^3 (solid squares) and 1151 kg/m^3 (open squares).

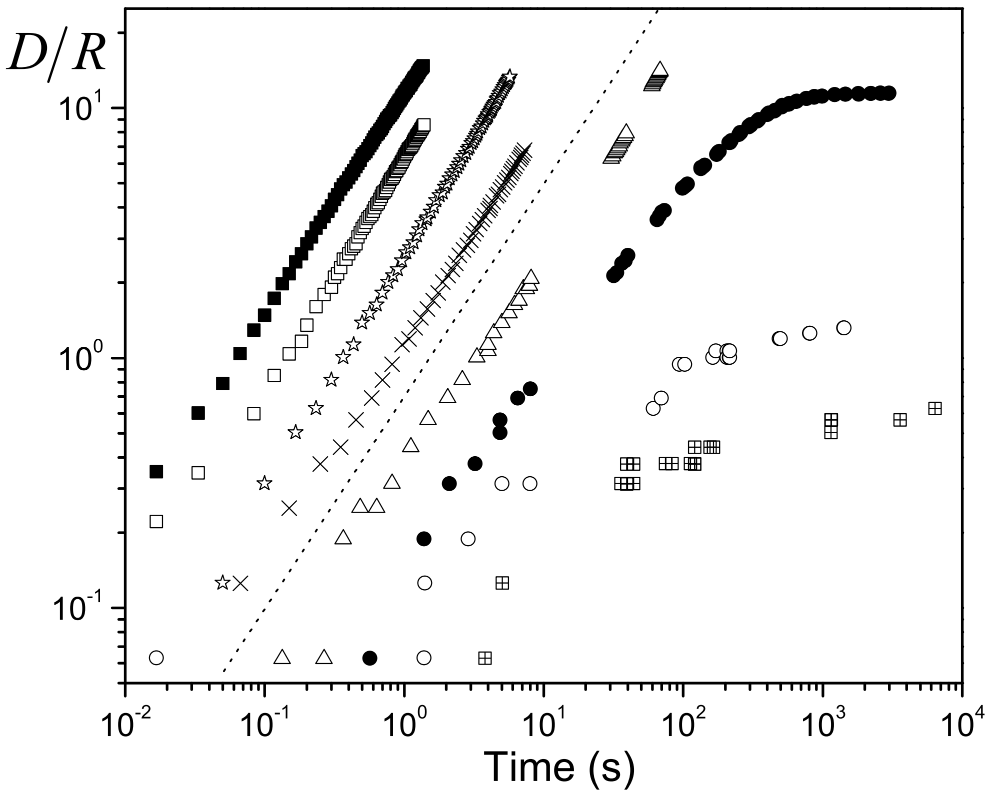

FIG. 2: D/R as a function of t for $\Delta\rho = 1345$ kg/m^3 and, from left to right, $t_w = 1$, 2, 5, 10, 20, 30, 40, and 45 min. The dotted line has a logarithmic slope of 1.

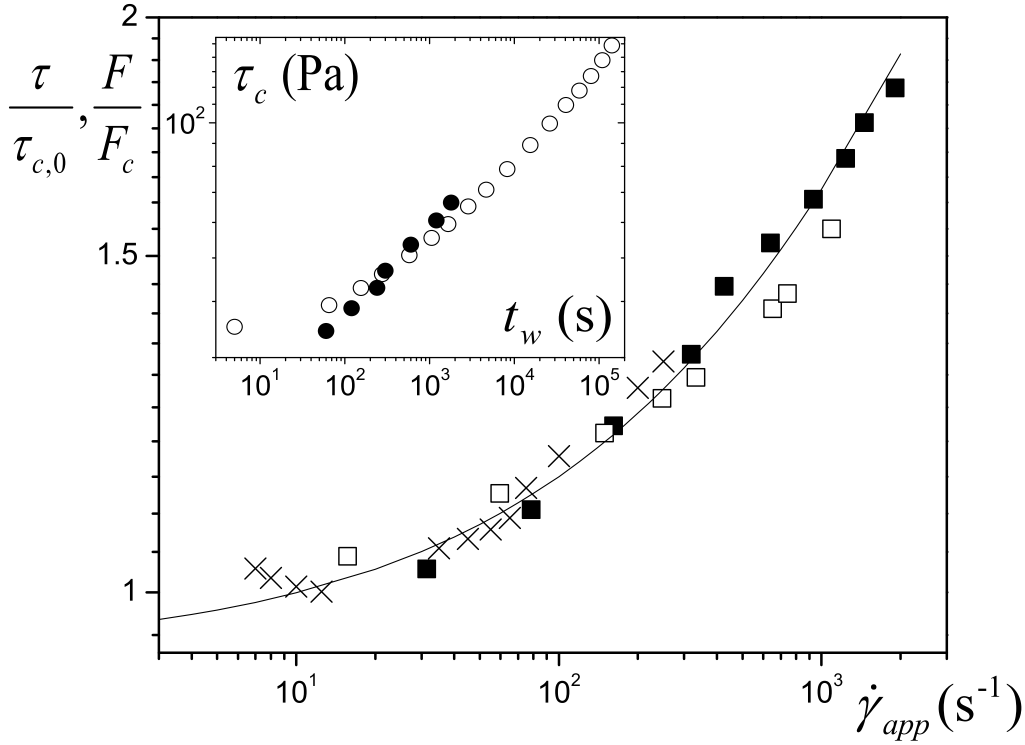

FIG. 3: Crosses: $\tau/\tau_{c,0}$ as a function of $\dot{\gamma}_{app}$ for the Laponite suspension in simple shear. Squares: F/F_c vs. v/ℓ obtained from the falling-sphere experiments with $t_w = 4$ min (solid squares) and 20 min (open squares). The curve is a fit of Eq. 4 to the force data. The inset shows the increase of the apparent yield stress τ_c with age determined from $G'(t_w)$ (open circles) and $F_c(t_w)$ (solid circles). The values of k_c and ℓ used in this analysis are discussed in the text.

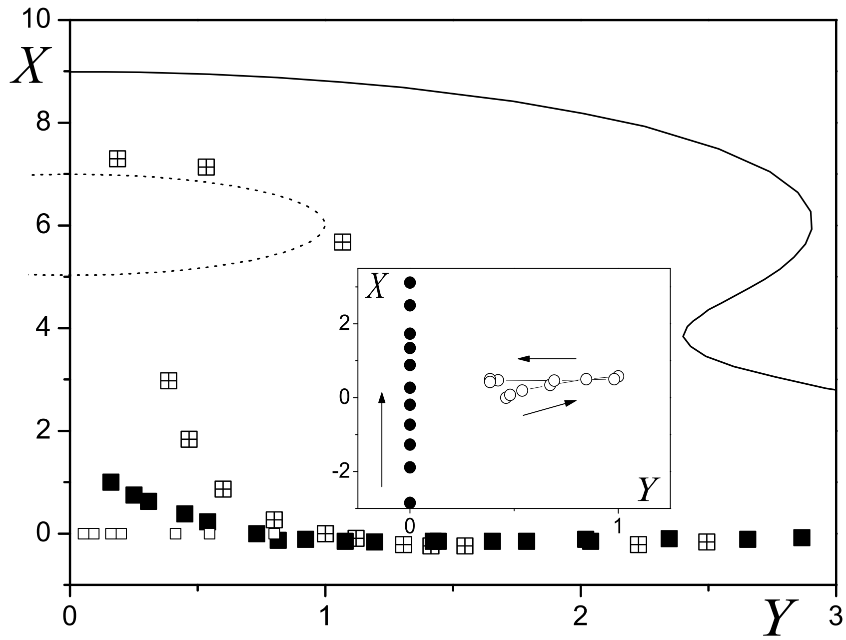

FIG. 4: Displacement X of a line of tracer particles initially at $X = 0$ after the sphere moves from $X = -3$ to $X = 6$, plotted against the distance Y from the axis of the sphere. X and Y are in units of R. Solid squares: a Laponite suspension similar to that described in the text; G' is 1.5 times larger due to two months additional storage prior to use. Here $R = 1.26$ cm and $v/R = 1.5$ s^{-1}. Crossed squares: Carbopol, a non-aging yield-stress fluid [3]; $R = 1.96$ cm and $v/R = 0.57$ s^{-1}. Open squares: the initial positions of the first seven of the particles from the Carbopol experiment, shown to emphasize that lateral displacements are significant in this case. Solid line: calculated displacements for a Newtonian fluid with no wall effects; here the results do not depend on v. The final position of the sphere is shown by the dotted line. The inset shows the position of a particular tracer particle at successive times (open circles) and the corresponding locations of the sphere's center (solid circles).